TO DEVELOP A NEW MINERAL CARBONATION PROCESS THAT HAVE A HIGH EFFICIENCY IN CO_2 ABSORPTION INTO INDUSTRY SLAG USING LOW ENERGY MECHANICAL MILLING

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"I hereby declare that the work in this project report is my own except for quotations and summaries which have been duly acknowledged.

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DEDICATION

To my beloved family, I am dedicated my thesis to all of you as my appreciation because always support me through my journey.

- Selamat Bin Hijor
- Saniah Binti Hj Shahri
- Mohd Fitrie Bin Jaafar
- Mohd Wazir Bin Selamat
- Siti Harah Binti Selamat
- Siti Hajar Binti Selamat
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ABSTRACT

Increase in the CO_2 emission in atmosphere due to the combustion of fossil fuels has caused serious global warming. Electricity generation, tranportation, and industrial waste are the main sectors indentified to contribute to the emission of CO_2 in Malaysia. In dealing with this issue, the absorption of CO_2 into industrial waste was experimentally studied by the utilization of mechanical grinding method. This research is to aim a development of new mineral carbonation process that has a high efficiency in the capture and storage of CO_2 with low energy consumption. In the first stage of this study, the behavior of CO_2 absorption on electric arc furnace and ladle furnace slag was studied by low energy mechanical milling It was found that the absorption is occured during milling. CO_2 was stored into the slag mainly as $CaCO_3$. Thus this indicates that the CO_2 can be stored permanently inside the slag with this method. In the next stage, the effect of dissolution of metal element into water on the behavior of CO_2 absorption was investigated by leaching test experiment. It was found that , concentration value of Fe in pure water is higher but in river water the concentration is lower, the dissolve concentration decreased with the increased in the number of the leaching time. Concentration will be increased at the earlier stage before it decreased at final of concentration. This case because the liquid became saturated and cannot be to dissolved. After the pH steeply increased gradually at an early stage of the elution of slag, it slightly decreased. The pH decreased with the increased in the number of elution. The changes of pH in leaching test it seemed to depend on the content of CaO in the slag. For mechanism of CO_2 absorption, morphological change of slag were study and the slag were characteried by using XRD, FE-SEM, and EDS.

ABSTRAK

Peningkatan pelepasan CO_2 di atmosfera akibat pembakaran bahan bakar fosil telah menyebabkan pemanasan global yang serius. Penjanaan elektrik, pengangkutan, dan sisa industri adalah sektor utama yang ditunjukkan untuk menyumbang kepada pelepasan CO_2 di Malaysia. Dalam menangani masalah ini, penyerapan CO_2 ke dalam sisa industri dikaji secara eksperimen dengan menggunakan kaedah pengisaran mekanikal. Penyelidikan ini bertujuan untuk pengembangan proses karbonasi mineral baru yang mempunyai kecekapan tinggi dalam penangkapan dan penyimpanan CO_2 dengan penggunaan tenaga yang rendah. Pada peringkat pertama kajian ini, tingkah laku penyerapan CO_2 pada tungku arka elektrik sanga dan sanga relau dikaji oleh penggilingan mekanikal tenaga rendah. Didapati bahawa penyerapan berlaku semasa penggilingan. CO_2 disimpan ke dalam terak terutamanya sebagai $CaCO_3$. Oleh itu, ini menunjukkan bahawa CO_2 dapat disimpan secara kekal di dalam terak dengan kaedah ini. Pada peringkat seterusnya, kesan pembubaran unsur logam ke dalam air terhadap tingkah laku penyerapan CO_2 disiasat dengan percubaan uji pencucian. Didapati bahawa, nilai kepekatan Fe dalam air tulen lebih tinggi tetapi dalam air sungai kepekatannya lebih rendah, kepekatan larut menurun dengan peningkatan jumlah masa pencucian. Kepekatan akan meningkat pada tahap awal sebelum penurunan pada akhir kepekatan. Ini berlaku kerana cecair menjadi tepu dan tidak dapat dilarutkan. Setelah pH meningkat secara beransur-ansur secara bertahap pada tahap awal pencairan sanga, sedikit menurun. PH menurun dengan peningkatan jumlah elusi. Perubahan pH dalam ujian pencucian sepertinya bergantung pada kandungan CaO dalam sanga. Untuk mekanisme penyerapan CO_2, perubahan morfologi sanga dikaji dan sanga ditandai dengan menggunakan XRD, FE-SEM, dan EDS.

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LIST OF ABBREVIATION AND SYMBOLS

ϴ	:	angle
ml	:	millimetre
l	:	litre
kg	:	kilogram
g	:	gram
mg/l	:	milligram per litre (concentration of liquid)
µm	:	micron meter
rpm	:	rotation per minute
Hz	:	Unit for amplitude
pH	:	measurement for acidic and alkaline
%	:	percentage
ASTM	:	American Society for Testing Materials
BOF	:	Basic Oxygen-Furnace
EAF	:	Electric Arc Furnace
LF	:	Ladle furnace slag
JIS	:	Japan Industrial Standard
EQS	:	Environment Quality Standards
OSHA	:	Occupational Safety and Health Administration
EPA	:	Protection Agency
SEM	:	Scanning Electron Microscopy
EDS	:	Energy Dispersive Spectroscopy
XRD	:	X-Ray Diffraction Analysis

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Carbon dioxide (CO_2) is one of the greenhouse gases (GHG) and the amount is much more higher than other GHG gases. Based on the prediction, about 285.73 million tonne will be emitted in year 2020 with the main contributors are from power generation, manufacturing industries, transportation and residential sector [1]. One of the promising technologies for the mitigation of CO_2 in the atmosphere is carbon capture and storage (CCS).

There are several other CCS technologies namely biological and geological storage, forestation and oil reservoirs, however mineral carbonation especially in industrial waste is still have some advantage in term of storage stability, long term continuous monitoring not required, and economically feasible [2]. While other mineral carbonation methods such as heat treatment, aging and aqueous carbonation requires high energy, long reaction time and use acidic solution. Carbonation through mechanical milling is a simple, use non-hazardous solution and inexpensive method that make it very attractive for CCS technology [3] However, the problem with suitable industrial waste for this application in Malaysia must be investigated.

The main objective of this research is To Develop A New Mineral Carbonation Process that have a high efficiency in CO_2 absorption into industry slag using Low Energy Mechanical Milling Consumption. The steel slag is collected from steelmaking industries in Malaysia and characterized to investigate the chemical composition. Then the slag is ground using low energy mechanical milling with specified parameters. In this research, the electric arc furnace (EAF) slag and ladle furnace (LF) slag is mainly used as a material for the carbonation process. The electric arc furnace slag and ladle furnace slag is an industrial residue that was discharge from steelmaking industry [4]. In using the mineral carbonation technique, there are also several potential materials that can be used as a sorbent. These materials are classified into minerals and waste materials. In this research, waste concrete also used as a sorbent to study the potential of reaction between waste concrete and carbon dioxide.

1.2 Problem Statement

Climate change, greenhouse gas effect and air pollution because of CO_2 emission has become major scientific issue and political issues. Many countries including Malaysia are playing an active-role in reducing CO_2 emission through national mitigation and intergovernmental mechanism such as The United Nations Framework Convention on Climate Change (UNFCC). Electricity generation, transportation, industrial, and residential are the main sectors identified to contribute to the emission of CO_2 in Malaysia [5] .

In comparison to other CCS technologies, mineral carbonation have some advantages in term of storage stability, long term continuous monitoring not required, and economically feasible [6]. Under the mineral carbonation, there are several techniques such as heat treatment, aging, aqueous carbonation and mechanical carbonation [7]. Compared to the first three techniques which require high energy, long reaction time and use acidic solution, carbonation through mechanical milling is simple, use non-hazardous solution, and inexpensive method that make it very attractive for CCS technology [8]. However, the problem with existing mechanical milling and devices is even though the energy required is still lower than that using heat treatment, but the energy consumed still lead to the low net amount of CO_2 absorption [9]. Thus, the use of low energy mechanical milling will be an effective solution for this problem.

In term of material used for the carbonation, carbonation can be done either using natural resources or industrial waste. However reutilization of natural resources such as olivine's, mafic, peridotite, wollastnite, and some others can contribute to some other environmental and public acceptance issues. Thus, the use of industrial waste is very much preferable in CCS [10]. Electric arc furnace and Ladle furnace are selected in this research because the main chemical composition of the slags contained high CaO that effective to absorp CO_2. Since the reutilization of steel slag in Malaysia is more focusing on the construction sector [11], the reutilization of steel slag in carbon capture and storage technology using low energy mechanical milling is very much important.

In this research, we are more focusing to develop a new mineral carbonation process that have a high efficiency in CO_2 absorption into industry slag using Low Energy Mechanical Milling Consumption. Since we are using a by-product from steel industry in which is steel slag , this study will not only contributing to the potential of CO_2 reduction from atmosphere, but also to the alternative application of the steel slag as sorbent for CO_2.

1.3 Objective

The objective of this research is to develop a new mineral carbonation process that have a high efficiency in CO_2 absorption with low energy consumption. In order to achieve this main objective, we have set these following sub objectives:

- To study the effect of milling on the behaviour of CO_2 absorption on EAF and LF steel slag in Malaysia.
- To investigate the dissolution behaviour of metal element from EAF and LF slag into water.
- To investigate the mechanism of the CO_2 sequestration into the steel slag.

1.4 Scope of the Study

The scope of the study as shown as follow:

- The slag is from electric arc furnace slag, ladle furnace slag and cement as waste concrete.
- The amount of CO_2 absorption is calculated based on the pressure change as follows: $nCO_2\ [mol] = \Delta\, PV/RT$

  Where ΔP is the amount of pressure change during stirring (Pa), V is the volume of the measuring system (m^3), R is the gas constant (8.3144598 $J\ mol^{-1}\ K^{-1}$), and T is room temperature (K).

- The milled ground carbonated slag was characterized using XRD and SEM/EDX. The liquid residue is observed using ICP-MS to investigate the concentration of metal elements dissolved in the water during the stirring. It is used to investigate the mechanism of the sequestration.
- The mechanism of CO_2 sequestration is investigated using the observation of pressure changed and the log C-pH diagram, where C is the concentration of dissolved metal elements inside the slag into water.

1.5 Significant of the Study

Global warming is expecting to turn worse and worse year by year. Since the main source of the global warming is the excessive emission of CO_2 gas into atmosphere, a technology to mitigate the emitted gas is very essential. It will contribute to the protection of society from any deadly effect of global warming in Malaysia such as sudden changes in the pattern of rainfalls, increase in the sea level at certain areas, increase in the number of deadly diseases likes dengue, and unpredictable occurrence of drought. Malaysian Journal of Environmental Management have conducted a research on global climate change and its effects on human habitat and environmental in Malaysia. They have reported that an analysis of temperature records in Malaysia shows a warming trend. For Malaysia, the temperature changes range from +0.70° C to +2.60° C, while precipitation changes ranges from -30% to +30%. Climate change in Malaysia showed this phenomenon have negative impacts on human habitats such as agriculture, forests, water resources, coastal resources, health and energy sector [12].

Thus study New Mineral Carbonation Process that have a high efficiency in CO_2 absorption into industry slag using Low Energy Mechanical Milling Consumption is very much important. It can contribute to the potential reduction of CO_2 sequestration in atmosphere. This will lead to the improvement of the environmental quality. Good environmental and air quality will lead the economic chaos is unlikely to happen and it will keep the economy to a stable and firm situation. The technology created can also be further developed for the utilization in industrial sites especially in manufacturing and power generation industries.

CHAPTER 2

LITERATURE REVIEW

2.1 CO_2 Emission in Malaysia

Malaysia is a Southeast Asian Country occupying parts of the Malay Peninsula and the island of Borneo. Figure 2.1 shows The Malaysia population from year 1950 to 2020. The current population of Malaysia is 32,365,999 as of January 25, 2020, based on the latest United Nations estimates. Malaysia population is equivalent to 0.42% of the total world population. [13].

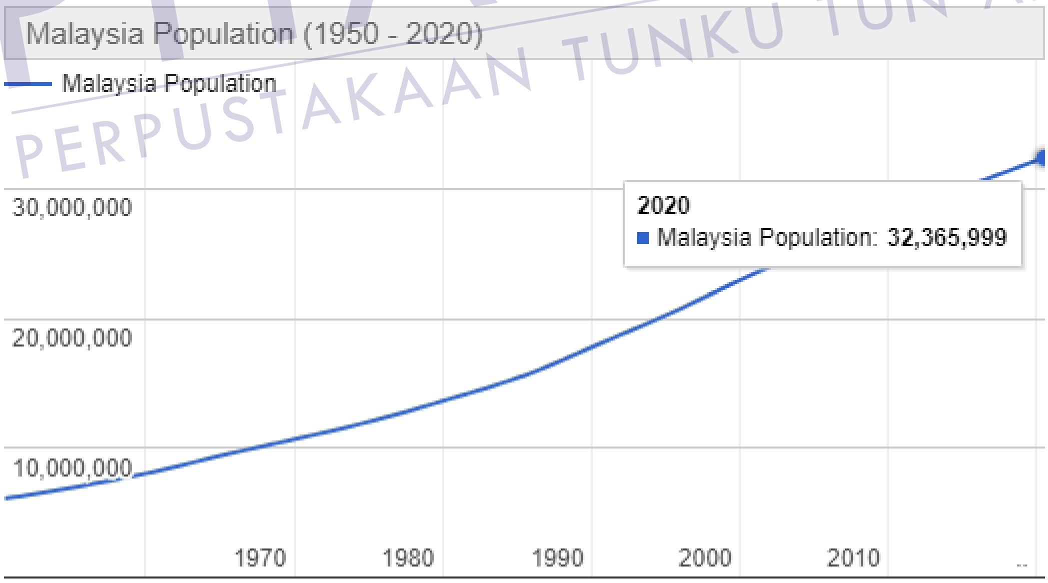

Figure 2.1: Malaysia population 1950 – 2020 [14]

In Malaysia, transportation system, electricity generation, industrial sectors and residual have been mentioned as the main contributors of CO_2 emission. According to statistics from Asian Pacific Energy Centre (APEC) [15], the outlook period for CO_2 emission from energy consumers in Malaysia is projected to grow about 4.2 annually reaching 414 million tone of dioxide carbon in 2030. Apart from fuel combustion, agricultural activities and waste materials includes disposal and water treatment are the other resources of GHGs generation in Malaysia. Based on Long-range Energy Alternatives Planning system (LEAP), the rate of CO_2 emission from fossil fuel-based sectors for the period of 2000 to 2020 can be model [16]. The structure of LEAP model which consist of energy demand, energy conversion, transmission, distribution, and end-use has been shown in Figure 2.2 schematically [17].

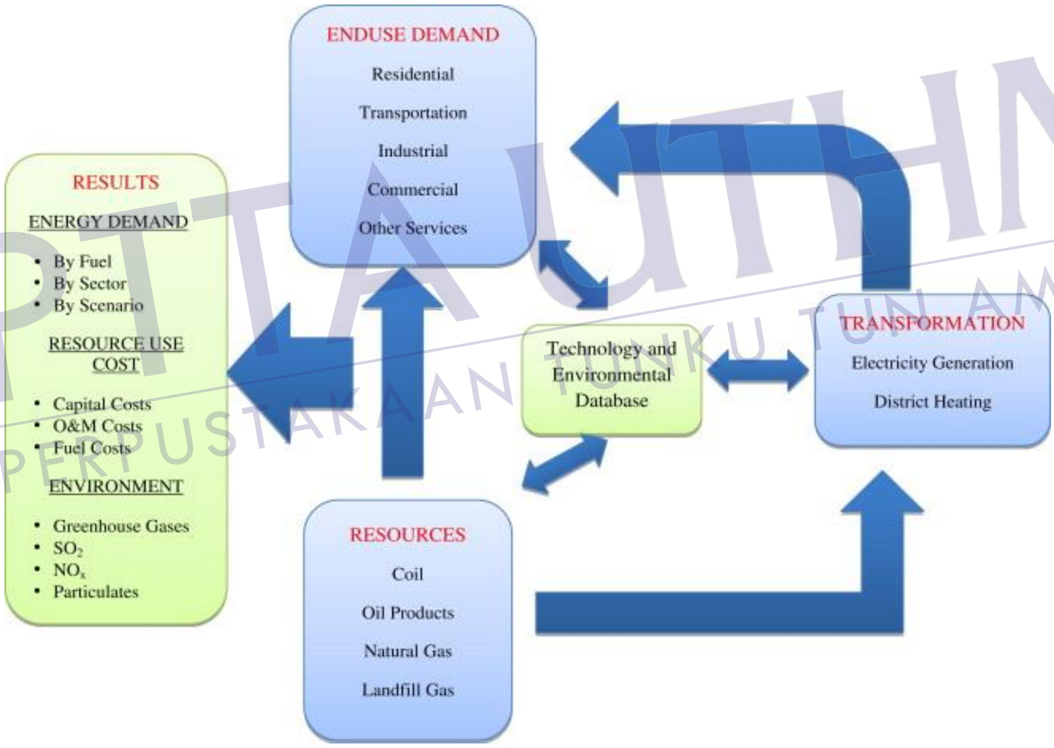

Figure 2.2: The schematic of LEAP model and economic assessment of landfill gas electricity generation in Korea using LEAP model [18].

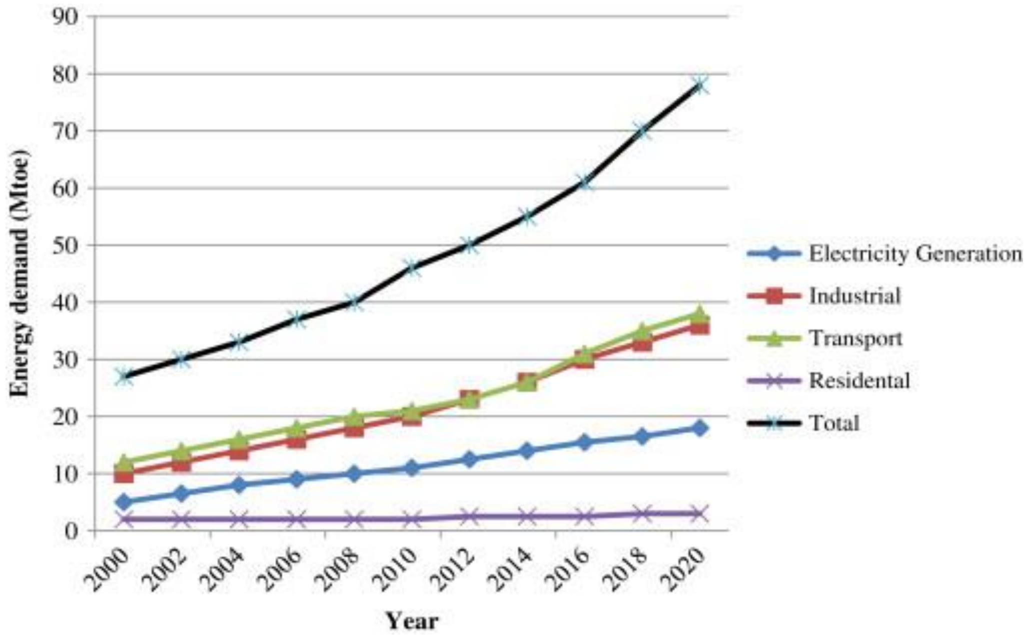

Figure 2.3: Energy demand from different sector in Malaysia [19]

Invoking to LEAP model and the related formulas [19], the trend of energy demand and CO_2 emission in Malaysia from 2000 to 2020 has been plotted in Figure 2.3 and Figure 2.4, respectively [20].

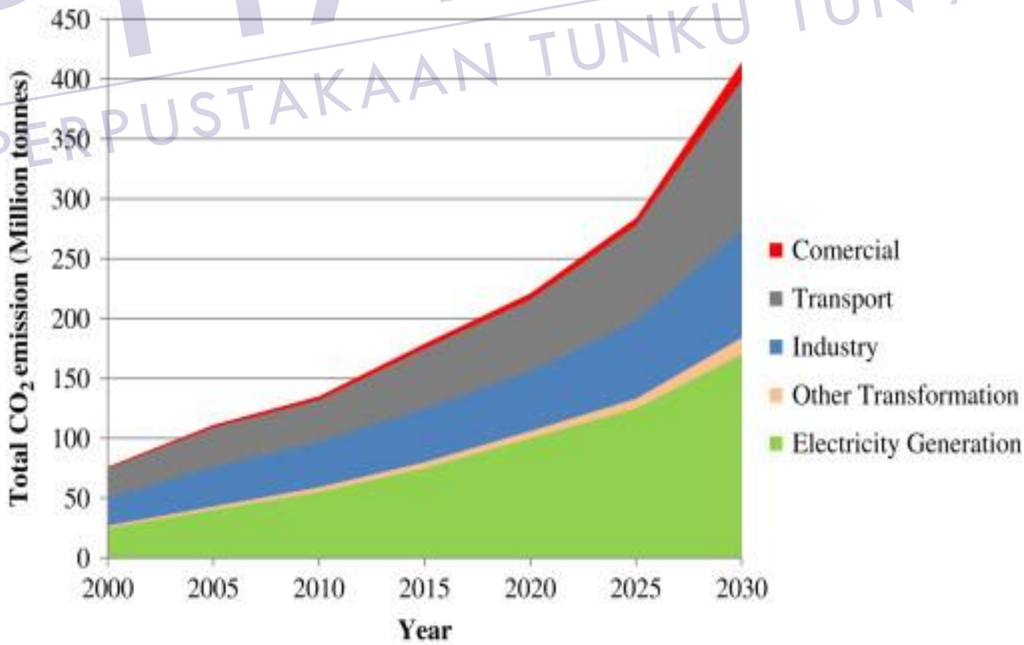

Figure 2.4: CO_2 emission from different emission in Malaysia [20]

The Gross Domestic Product per capita in Malaysia was last recorded at 11028.20 US dollars in 2016. The GDP per Capita in Malaysia is equivalent to 87 percent of the world's average. GDP per capita in Malaysia averaged 5046.70 USD from 1960 until 2016, reaching an all-time high of 11028.20 USD in 2016 and a record low of 1408.60 USD in 1960.

The trend of gross domestic product per capita in Malaysia is shown is Figure 2.5. Gross domestic product (GDP) contribute the ratio of greenhouse gas emission. An emission intensity is the average emission rate of a given pollutant [21] from a given source relative to the intensity of a specific activity. Emission Intensity of economic is a measure of greenhouse gas emission per unit of economic activity, usually measured in GDP. Unlike absolute emission value which measures the quantum of emission, emission intensity is a relative value with respect to GDP.

Reduction of emission intensity of GDP means reducing pollution per unit of GDP. But if GDP grows, the emission also grow along with it. For instance, if GDP of a Malaysia remain constant, the reduction in values of emission intensity reduces the pollution proportionately. But if the GDP growth rate increase, then the overall reduction in value of absolute emission may not reduce.

Figure 2.5: Malaysia GDP Per capita [21]

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